

THAT WHICH IS CLAIMED:

1. A method of producing nitrogen trifluoride, comprising:  
providing a fluorine-containing feed stream;  
5                   contacting the fluorine-containing feed stream with liquid ammonium  
acid fluoride in a reaction zone for a time and under conditions sufficient to produce  
nitrogen trifluoride,  
decreasing the effective melt acidity value of the liquid ammonium  
acid fluoride during said contacting step; and  
10                   removing a reaction product stream comprising nitrogen trifluoride  
from the reaction zone.
2. A method according to Claim 1, wherein said contacting step comprises  
contacting the fluorine-containing feed stream with liquid ammonium acid fluoride in  
a series of reactors, wherein each successive reactor contains ammonium acid fluoride  
having a progressively lower bulk melt acidity value.
3. A method according to Claim 1, wherein said decreasing step comprises  
decreasing the effective melt acidity value of the liquid ammonium acid fluoride from  
a value above the optimum value resulting in the highest nitrogen trifluoride yield at  
reaction zone temperature and pressure to approximately the optimum value.
4. A method according to Claim 1, wherein the fluorine-containing feed  
stream comprises elemental fluorine and hydrogen fluoride.
5. A method according to Claim 1, wherein the ammonium acid fluoride has  
an acid-base stoichiometry of  $\text{NH}_4\text{M}_y\text{F}_z(\text{HF})_x$ , wherein M is a metal selected from the  
group consisting of Group IA through VA, Group IB through VIIB and Group VIII of  
the Periodic Table of Elements or mixtures thereof; y is 0-12; z is 1-12; and x is the  
5                   melt acidity value.

6. A method according to Claim 1, wherein the ammonium acid fluoride has an acid-base stoichiometry of  $\text{NH}_4\text{F}(\text{HF})_x$ , wherein x is the melt acidity value.

7. A method of producing nitrogen trifluoride, comprising:

providing a gaseous mixture of elemental fluorine and hydrogen fluoride;

feeding the gaseous mixture into a reaction zone containing a bulk ammonium acid fluoride;

contacting the gaseous mixture with the bulk liquid ammonium acid fluoride for a time and under conditions sufficient to produce nitrogen trifluoride, wherein the initial effective melt acidity value of the ammonium acid fluoride is greater than the melt acidity value of the bulk liquid ammonium acid fluoride; and  
removing a reaction product stream comprising nitrogen trifluoride from the reaction zone.

8. A method according to Claim 7, wherein the initial effective melt acidity value is at least about 0.05 greater than the melt acidity value of the bulk liquid ammonium acid fluoride in the reaction zone.

9. A method according to Claim 7, wherein the bulk liquid ammonium acid fluoride melt acidity value is less than about 1.8.

10. A method according to Claim 7, wherein the bulk liquid ammonium acid fluoride melt acidity value is less than about 1.6.

11. A method according to Claim 7, wherein the bulk liquid ammonium acid fluoride melt acidity value is less than about 1.5.

12. A method according to Claim 7, wherein the initial partial pressure of hydrogen fluoride in the gaseous mixture is at least about 15 kPa at the operating temperature and pressure of the reaction zone.

13. A method according to Claim 7, wherein the initial partial pressure of hydrogen fluoride in the gaseous mixture is at least about 25 kPa at the operating temperature and pressure of the reaction zone.

14. A method according to Claim 7, wherein the initial partial pressure of hydrogen fluoride in the gaseous mixture is at least about 40 kPa at the operating temperature and pressure of the reaction zone.

15. A method according to Claim 7, wherein the operating temperature of the reaction zone is about 120 to about 150°C.

16. A method according to Claim 7, wherein the operating pressure of the reaction zone is about 80 to about 200 kPa.

17. A method according to Claim 7, wherein said contacting step occurs in a stirred tank reactor.

18. A method of producing nitrogen trifluoride, comprising:  
providing a gaseous mixture of elemental fluorine and hydrogen fluoride;

feeding the gaseous mixture into a reaction zone containing a bulk ammonium acid fluoride;

contacting the gaseous mixture with the bulk liquid ammonium acid fluoride having a bulk melt acidity value of less than about 2.0 for a time and under conditions sufficient to produce nitrogen trifluoride, wherein the initial effective melt acidity value is greater than the melt acidity value of the bulk liquid ammonium acid fluoride, and further wherein the initial partial pressure of hydrogen fluoride in the gaseous mixture is at least about 15 kPa at the operating temperature and pressure of the reaction zone;

removing a reaction product stream from the reaction zone, the reaction product stream comprising nitrogen trifluoride and entrained liquid ammonium acid fluoride;

introducing the reaction product stream into a regeneration zone,  
wherein the operating pressure of the regeneration zone is lower than the operating  
pressure of the reaction zone such that gaseous hydrogen fluoride is released from the  
entrained liquid ammonium acid fluoride;

5                    removing a regeneration product stream from the regeneration zone,  
the regeneration product stream comprising nitrogen trifluoride and hydrogen  
fluoride;

introducing the regeneration product stream into a separation zone to  
separate hydrogen fluoride from nitrogen trifluoride;

10                   recycling liquid ammonium acid fluoride from the regeneration zone to  
the reaction zone; and

recycling at least a portion of the hydrogen fluoride separated in the  
separation zone for use in the gaseous mixture of elemental fluorine and hydrogen  
fluoride.

19. A method according to Claim 18, wherein the initial effective melt acidity  
value is at least about 0.05 greater than the melt acidity value of the bulk liquid  
ammonium acid fluoride in the reaction zone.

20. A method according to Claim 18, wherein the bulk liquid ammonium acid  
fluoride melt acidity value in the reaction zone is less than about 1.8.

21. A method according to Claim 18, wherein the bulk liquid ammonium acid  
fluoride melt acidity value in the reaction zone is less than about 1.6.

22. A method according to Claim 18, wherein the bulk liquid ammonium acid  
fluoride melt acidity value in the reaction zone is less than about 1.5.

23. A method according to Claim 18, wherein the initial partial pressure of  
hydrogen fluoride in the gaseous mixture is at least about 25 kPa at the operating  
temperature of the reaction zone.

24. A method according to Claim 18, wherein the initial partial pressure of hydrogen fluoride in the gaseous mixture is at least about 40 kPa at the operating temperature and pressure of the reaction zone.

25. A method according to Claim 18, wherein the operating temperature of the reaction zone is about 120 to about 150°C.

26. A method according to Claim 18, wherein the operating pressure of the reaction zone is about 80 to about 200 kPa.

27. A method according to Claim 18, wherein the reaction zone and the regeneration zone are located within separate stirred tank reactors.

28. A method according to Claim 18, wherein the operating pressure of the reaction zone is at least about 50 kPa higher than the operating pressure of the regeneration zone.

29. A method according to Claim 18, wherein the operating pressure of the regeneration zone is about 5 to about 20 kPa.

30. A method according to Claim 18, wherein the regeneration zone is located at an elevation at least about 6 meters higher than the elevation of the reaction zone.

31. A method according to Claim 18, wherein said step of recycling liquid ammonium acid fluoride from the regeneration zone to the reaction zone comprises:

5        passing liquid ammonium acid fluoride from the regeneration zone to a gas-liquid separation tank in order to separate a gas phase from the liquid ammonium acid fluoride;

             combining the gas phase produced in the gas-liquid separation tank with the regeneration product stream; and

             passing liquid ammonium acid fluoride from the separation tank to the reaction zone.

32. A method according to Claim 18, wherein the recycled liquid ammonium acid fluoride from the regeneration zone is mixed with the gaseous mixture of elemental fluorine and hydrogen fluoride prior to entry into the reaction zone.

33. A method according to Claim 18, wherein the flow rate of the recycled liquid ammonium acid fluoride entering the reaction zone is at least about 1000 times the stoichiometric flow rate.

34. A method according to Claim 33, wherein the flow rate of the recycled liquid ammonium acid fluoride entering the reaction zone is at least about 2000 times the stoichiometric flow rate.

35. A method according to Claim 18, further comprising:  
reacting ammonia with hydrogen fluoride in a second reaction zone under conditions sufficient to produce ammonium acid fluoride;  
removing a liquid ammonium acid fluoride product stream from the  
5 second reaction zone; and  
introducing the ammonium acid fluoride product stream into the regeneration zone.

36. A method according to Claim 35, further comprising:  
contacting the liquid ammonium acid fluoride product stream from the second reaction zone with the regeneration product stream from the regeneration zone in order to remove entrained ammonium acid fluoride from the regeneration product  
5 stream.

37. A method according to claim 36, wherein said step of contacting the ammonium acid fluoride product stream from the second reaction zone with the regeneration product stream from the regeneration zone comprises contacting the ammonium acid fluoride product stream and the regeneration product stream in a  
5 demister.

38. A method according to Claim 35, wherein at least a portion of the hydrogen fluoride separated in the separation zone is recycled for reaction with the ammonia in the second reaction zone.

39. A method according to Claim 18, wherein at least a portion of the hydrogen fluoride separated in the separation zone is collected in a byproduct stream.

40. An apparatus for producing nitrogen trifluoride, comprising:  
a gaseous mixture supply comprising elemental fluorine and hydrogen fluoride; and  
a first reactor in fluid communication with said gaseous mixture supply and comprising a reaction zone and an outlet, wherein said reaction zone is operatively positioned to contact said gaseous mixture with a bulk liquid ammonium acid fluoride.

41. An apparatus according to Claim 40, further comprising:  
a regenerator in fluid communication with the outlet of said first reactor and comprising a regeneration zone and a product outlet, wherein said regeneration zone is operatively positioned to separate gaseous hydrogen fluoride from liquid ammonium acid fluoride, and wherein said product outlet is operatively positioned to remove a regeneration product stream from said regeneration zone; and  
a separator in fluid communication with said product outlet of said regenerator and operatively positioned to separate hydrogen fluoride from nitrogen trifluoride, said separator comprising a hydrogen fluoride outlet and a nitrogen trifluoride outlet.

42. An apparatus according to Claim 41, wherein said gaseous mixture supply is in fluid communication with said hydrogen fluoride outlet of said separator.

43. An apparatus according to Claim 41, wherein said separator comprises a gas-liquid separation tank having a gas phase outlet and a liquid outlet and a reflux condenser in fluid communication with said gas phase outlet of said separation tank.

44. An apparatus according to Claim 41, wherein said regenerator further comprises a recycle outlet in fluid communication with said first reactor and operatively positioned to recycle liquid ammonium acid fluoride to said first reactor.

45. An apparatus according to Claim 41, further comprising a gas-liquid separation tank in fluid communication with said recycle outlet of said regenerator and operatively positioned to separate a gas phase from the liquid ammonium acid fluoride prior to recycle to said first reactor.

46. An apparatus according to Claim 41, further comprising a demister in fluid communication with said product outlet of said regenerator and operatively positioned to remove entrained liquid ammonium acid fluoride from the regeneration product stream.

47. An apparatus according to Claim 41, further comprising:  
an ammonia supply; and  
an ammonium acid fluoride reactor in fluid communication with said ammonia supply and said hydrogen fluoride outlet of said separator and operatively  
5 positioned to react ammonia and hydrogen fluoride to form ammonium acid fluoride, wherein said ammonium acid fluoride reactor comprises an outlet in fluid communication with said regenerator.

48. An apparatus according to Claim 41, further comprising:  
a demister in fluid communication with said product outlet of said regenerator and operatively positioned to remove entrained liquid ammonium acid fluoride from the gaseous product stream;  
5 an ammonia supply; and  
an ammonium acid fluoride reactor in fluid communication with said ammonia supply and said hydrogen fluoride outlet of said separator and operatively positioned to react ammonia and hydrogen fluoride to form ammonium acid fluoride, wherein said ammonium acid fluoride reactor comprises an outlet in fluid  
10 communication with said demister.



49. An apparatus for producing nitrogen trifluoride, comprising:
- a gaseous mixture supply comprising elemental fluorine and hydrogen fluoride;
  - a first reactor in fluid communication with said gaseous mixture supply and comprising a reaction zone and an outlet, wherein said reaction zone is operatively positioned to contact said gaseous mixture with a bulk liquid ammonium acid fluoride having an acid-base stoichiometry of  $\text{NH}_4\text{F}(\text{HF})_x$ , wherein x is the melt acidity value;
  - a regenerator in fluid communication with the outlet of said first reactor and comprising a regeneration zone, a product outlet and a recycle outlet, wherein said regeneration zone is operatively positioned to separate hydrogen fluoride from liquid ammonium acid fluoride, wherein said regeneration product outlet is operatively positioned to remove a regeneration product stream from said regeneration zone, and wherein said recycle outlet is in fluid communication with said first reactor and operatively positioned to recycle liquid ammonium acid fluoride to said first reactor; and
  - a separator in fluid communication with said product outlet of said regenerator and operatively positioned to separate hydrogen fluoride from nitrogen trifluoride, said separator comprising a hydrogen fluoride outlet and a nitrogen trifluoride outlet, wherein said hydrogen fluoride outlet is in fluid communication with said gaseous mixture supply.